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## PATENT ABSTRACTS OF JAPAN

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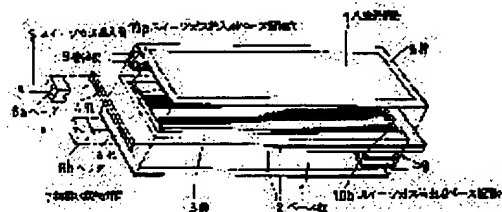
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## (54) PLATE TYPE HYDROGEN SEPARATION MEMBRANE MODULE

## (57)Abstract:

PROBLEM TO BE SOLVED: To improve both the durability (life) of a hydrogen separation membrane module and the production cost and reduce the production cost of the hydrogen separation membrane module.

SOLUTION: Plural grooves 3 in which a sweep gas for flowing hydrogen permeated from the outside flows are formed on both surfaces of base plates 2 and one or plural reinforcing plates 9 composed of a metallic porous material and membranes 1 and 1 capable of selectively permeating only the hydrogen are arranged on both surfaces of the base plates 2. Plural holes 4 are arranged in the center of the base plates so that the sweep gas and hydrogen may turn back at the ends thereof.



## LEGAL STATUS

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**CLAIMS**

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[Claim(s)]

[Claim 1] The monotonous mold hydrogen demarcation membrane module characterized by to have established two or more slots which pass the sweep gas for passing the hydrogen penetrated from the outside in both sides of a base plate, to have arranged in both sides of this base plate the transparency film which penetrates alternatively only the back up plate which consists of 1 or a metal porous body of two or more sheets, and hydrogen, and to prepare two or more holes in the core of a base plate so that the sweep gas and the hydrogen gas which flowed from the slot on double-sided may turn up and flow at the end.

[Claim 2] The monotonous mold hydrogen demarcation membrane module according to claim 1 characterized by the above-mentioned base plate joining two plates which carried out recessing to the both sides by etching.

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the monotonous mold hydrogen demarcation membrane module used for the manufacturing installation and refiner of high grade hydrogen gas in detail about a monotonous mold hydrogen demarcation membrane module.

[0002]

[Description of the Prior Art] As a means to separate and collect the hydrogen gas of the high grade from hydrogen mixed gas, the approach using the metal membrane which penetrates hydrogen alternatively is already put in practical use. Pd film method is used for the super-high grade hydrogen manufacturing installation in industry, such as semiconductor manufacture, as an industrial example of application. Moreover, application to the petrochemical industries, such as ammonia manufacture, is also progressing in recent years. There are the following approaches as the manufacture approach of Pd film.

\*\* Anneal and cold-roll hydrogen permselectivity metals, such as Pd and a Pd-Ag (palladium and silver) alloy.

\*\* Make the thin film of Pd or Pd alloy form in the front face of the porous body which turns into a base material of Pd film by plating or vacuum evaporation actuation (for example, thing adapting the vacuum deposition method by electroplating, electroless deposition, and electron beam heating etc.).

In order to use it industrially, a hydrogen demarcation membrane module is constituted combining a base plate with the space in which the hydrogen permeability film (foil) made with the above means is carried out outside, it puts on a hydrogen mixed-gas side, and the hydrogen to penetrate is brought together. Among both, the perforated plate with which film reinforcement is generally reinforced is inserted.

[0003] In addition, they may be the case where a base plate is monotonous in a modular configuration, and a cylinder. When the hydrogen permselective membrane by Pd alloy is explained as an example, Pd and its alloy film have the property to make hydrogen penetrate alternatively. It is explained that Pd film is adsorbed, the hydrogen content child in hydrogen mixed gas will be in an atomic condition, and the phenomenon is ionized further, is diffused and recombined with the membranous opposite side, and becomes hydrogen gas again. Recently, engine-performance improvement efforts of Pd film are paid, and the approach of blending various kinds of rare earth elements etc. is performed.

[0004] About the configuration of the conventional hydrogen demarcation membrane module, an example in case a base plate is flat sheet structure is explained based on drawing 3 and drawing 4. In addition, drawing 3 shows the modular example of a configuration, and drawing 4 shows a modular appearance. The hydrogen permselectivity foil 1 made from a Pd-Ag alloy manufactured with cold rolling is placed on the back up plate 9. The back up plate 9 is the thing in which much openings were formed, and it is used by two or more sheets in many cases so that it may increase reinforcement synthetically. In this example, the back up plate 9 of three sheets is arranged. The base plate 2 with the slot 3 for collecting the purification hydrogen for which the hydrogen permeability foil 1 and the back up plate 9 are passed, and it gathers is put on the bottom of it, a seal weld 11 is given to the perimeter of each part material (1, 9, and 2), the whole is unified, and it considers as a demarcation membrane module. In this example, the purification hydrogen gas gathering in a slot 3 is attached in the base plate 2, makes the flowing sweep gas installation tubing 6 to a slot 3, a steam, etc. flow, and samples purification hydrogen gas from the purified gas extraction tubing 7 attached in the side face which counters. In addition, tubing 6 and 7 is welded to the base plate 2, and is attached.

[0005]

[Problem(s) to be Solved by the Invention] By the way, generally the hydrogen demarcation membrane by which arrangement immobilization was carried out is used in a hydrogen decollator or a refiner under pressurization and an elevated-temperature ambient atmosphere (6-10atm, 400-700-degreeC). Therefore, in equipment operation, the body of

a hydrogen demarcation membrane module and its attachment piping are expanded at the time of start and stop and contracted by the temperature change. In order for thermal stress hysteresis to mainly join the weld zone of attachment piping, the fatigue defect occurred and the life of a hydrogen demarcation membrane module was contracted by what attached sweep gas (and hydrogen) piping in the conventional both ends of a hydrogen demarcation membrane module. Then, although the daily check (maintenance) was needed for the avoid accident, the exchange cost of the costs and a hydrogen demarcation membrane was great. Moreover, in operation accompanied by the start and stop of one day, the life of a hydrogen demarcation membrane module was 30 - 120 days. This invention was made in view of the above-mentioned actual condition, improves the durability (life) and manufacture cost of a hydrogen demarcation membrane module, and aims at aiming at the reduction.

[0006]

[Means for Solving the Problem] The back up plate which establishes two or more slots where the sweep gas for passing the hydrogen penetrated from the outside flows in both sides of a base plate, and becomes both sides of this base plate from 1 or the metal porous body of two or more sheets in this invention in order to attain the above-mentioned purpose, The transparency film which penetrates only hydrogen alternatively is arranged, two or more holes are further prepared in the core of a base plate, and it is considering as the configuration arranged so that the sweep gas and hydrogen gas which flowed from the slot on double-sided may turn up at the end and it may come to the above-mentioned feed hole. In this invention, generating of the thermal stress which joins a module is prevented by having considered as the above-mentioned configuration by arranging the gas inlet and extraction opening of a hydrogen demarcation membrane module on the same side face of this module. In addition, the manufacture cost of a base plate has the most advantageous etching method in respect of processing of a base plate and its cost of materials, when cutting a 2mm (width of face) x 1.5mm (depth) slot in consideration of allowable pressure loss, as a result of examining the machining method, precision casting, and the etching method.

[0007] Next, the configuration of the hydrogen demarcation membrane module which applied this invention is explained using drawing 1 and drawing 2 . Sweep gas is introduced from header 8a of an edge in the structure, and the base plates 2 and 2 make two or more holes 4 which flowed, carried out each two or more slots of both flat surfaces by return at the edge of the opposite side, and established them in the core the structure which flows and is discharged from another header 8b. A hydrogen division plate consists of a hydrogen permeability foil 1 which joined the back up plate 9 (a part is omitted and shown) which consists of a metal porous body of 1-3 sheets put on both sides of the base plate 2, and the film which penetrates only hydrogen alternatively, and is formed by attaching in one end of the base plates 2 and 2 with the gas installation tubing 6, and attaching Headers 8a and 8b in the gas exhaust pipe 7 by welding, respectively. Although the slot of the base plates 2 and 2 may be processed by which approaches, such as the machining method, precision casting, and etching, it makes a slot from etching, and it is divided into two or more sheets, it produces it, and the thing joined after that, especially the thing which joined two sheets which carried out recessing to both sides can produce it thinly cheaply.

[0008] The hydrogen demarcation membrane module which applied this invention is set centering on the base plate 2, and the hydrogen permeability film 1 is put on both the front face to the back up plate 9 and also its outside both sides. The sweep gas introduced into this module the hole 4 formed in the doubling section of two base plates 2 A passage, The inside of the hydrogen permeable film 1 of the base plates 2 and 2 of each opening and the slot 3 currently installed crosswise [ of this base plate ] from the edge of the base plates 2 and 2 through hole 10a which carried out opening to 1 side is flowed. Wash away the hydrogen which exudes inside a hydrogen permeable film 1 to an opposite end a sweep gas installation tubing anchoring-side, make it flow into the hole 4 side of the mating face of the base plates 2 and 2 from hole 10b of the edge of the base plates 2 and 2, and it goes back with the flow direction at the time of sweep gas installation. It is extracted out of a module from the purification hydrogen extraction tubing 7, and becomes product hydrogen. In addition, generally a steam is used for sweep gas.

[0009]

[Function] Since hydrogen mixed-gas (raw material) piping and purification hydrogen gas piping which are connected to a hydrogen demarcation membrane module are attached in the same side face, the end of a hydrogen demarcation membrane module turns into the free end, and operation of this film, and hydrogen gas installation and extraction tubing is possible in the condition of having been wide opened from heat telescopic motion with this invention. Therefore, the thermal stress in the piping anchoring weld zone to a membrane module is not generated, either, but the synthetic manufacture cost cut of a membrane module can be aimed at by having attached the hydrogen permeation foil in base \*\*\*\* external surface.

[0010]

[Example] First, the engine performance of a conventional method is outlined as an example of a comparison of the

effectiveness of this invention. (Example of a comparison) Although the processing yield did not exceed about 60% in the production process of the hydrogen demarcation membrane module using a conventional method by the cylinder method which needs bending, by the plate process formula shown in drawing 3 and drawing 4, the yield became about 90% and practicality has been improved. However, by the conventional monotonous base method, the fatigue failure accidents of the piping anchoring section occurred frequently during operation, and the trouble had arisen like usual. The result of having conducted the experiment which applies thermal stress to Pd foil hydrogen demarcation membrane module manufactured by the general technique in a laboratory repeatedly is shown below.

Experiment conditions: It took out, after heating a specimen to 600-degreeC with a heating furnace, and air cooling (1-hour maintenance after specimen temperature is stabilized, respectively) was carried out at the room temperature (about 30-degreeC).

Experimental result: Repeat count of a temperature change which resulted in defective generating of a weld zone 25 times.

In addition, the period of the time of the occurrence of the breakage accident of the tubing attachment section was presumed to be about 100 days. By this conventional example module, the base plate was made into SUS material and applied the machining method. the dimension of a base plate -- 70mm(W) x300mm(L) x -- it was 5mm (t). The gas installation and extraction tubing attached in the base plate offered as a sample are 100mm in SUS tubing and die length (L).

[0011] (Example) The hydrogen permeation foils 1 and 1 were attached in both sides which use SUS material as the base plate 2, the hydrogen demarcation membrane module of this invention was made as an experiment, and the system durability was predicted. The magnitude of a hydrogen demarcation membrane module presupposed that it is the same as the conventional example, and processing of the base plate 2 was performed by the chemical etching method. Base board thickness could be 5mm (thickness) in which double-sided processing of need slot dimension 2mm(width of face) x1.5mm (depth) is possible. Although the experiment was conducted on the same conditions as the aforementioned example of the conventional comparison, the count of a temperature change generating of a weld zone organization defect was accepted to be became 75 times, the durable days in the system were presumed to be about 300 days, and it has checked that improvement effectiveness was large. Although this hydrogen demarcation membrane module has set two base plates 2 and 2, a hydrogen permeability foil is in both sides, and the number of components is the same as that of two modules of a conventional type. However, since the thickness of the base plates 2 and 2 is small, module thickness becomes small and compact arrangement is attained. Module assembly cost becomes lower than the conventional method about 30% per film surface product.

[0012]

[Effect of the Invention] Since gas installation and extraction tubing are formed in the same side face of a hydrogen demarcation membrane module and piping does not bind a hydrogen demarcation membrane module in this invention like [ in case this anchoring tubing has been arranged to modular both ends ] also when a temperature change arises, there is no generating of thermal stress in a piping attachment weld zone. Therefore, the fatigue-breaking trouble by thermal stress generating in the piping anchoring section used as the technical problem in the hydrogen demarcation membrane module of a conventional type can be prevented. Furthermore, in this invention, a base plate manufacturing cost can be reduced about to 1/3 by applying the etching method to manufacture of a base plate compared with what applies the conventional machining method, and it is economically advantageous. Since it has a hydrogen permeable film in both sides by the hydrogen demarcation membrane module of this invention, the cost per unit membrane side becomes low, and a compact array within a real unit is more possible for it than the conventional method further again.

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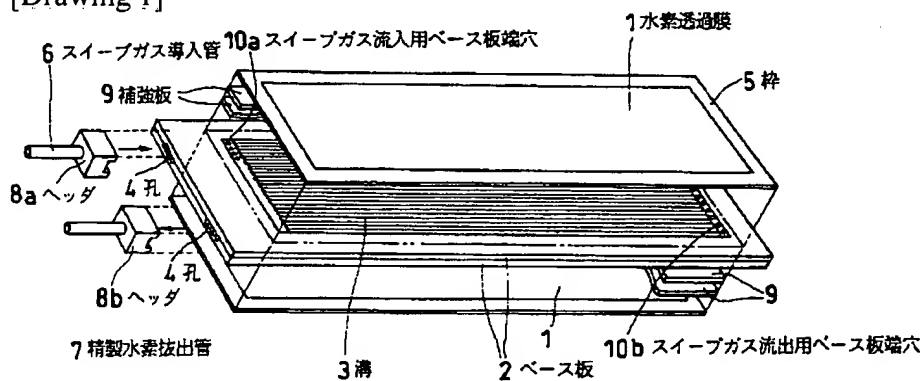
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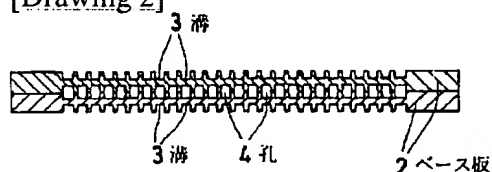
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## DRAWINGS

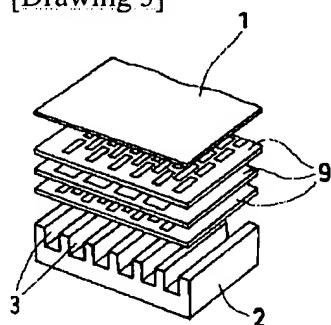
[Drawing 1]



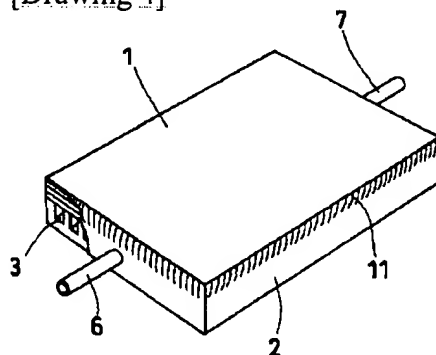
[Drawing 2]



[Drawing 3]



[Drawing 4]



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